

close to their source¹². Volcanism has been invoked to account for the particularly high proportion of natural emissions in Antarctic snow⁶.

A substantial natural input of trace metals to Greenland snow has been reported by other workers¹³, and a large enrichment of heavy metals in comparison with crustal reference elements has been firmly demonstrated in both ancient and modern polar snows^{13,14}. Until the mechanisms of trace metal deposition are clearly understood, any attempts to assess temporal changes in atmospheric loadings from evidence in ice cores must be tentative.

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NRIAGU REPLIES—Landy *et al.* have misinterpreted my report¹. I was careful to say changes in rates of trace metal deposition rather than the ratios in trace metal concentrations in the ice layers. My statement clearly pertains to dF/dt (where F is metal flux rate at each ice layer and t is time), and is quite different from the linear relation, C_1/C_2 (C_1, C_2 are metal concentrations of dated ice layers) assumed by Landy *et al.* The data tabulated by Landy *et al.* may thus bear no relationship whatsoever to the statement in my paper.

Landy *et al.* have dismissed the data of Jaworowski *et al.*² because "it contained

insufficient information about local geology and analytical procedures". However, more than a page of this paper was devoted to materials and methods and included the cogent reference to the work on the characteristics of the Storebreen glacier. The high values of metals reported may not be due to sample contamination as such but may instead be related to deposition of long-range transported material from industrial centres of Europe (such as sulphate deposition and acid rains, see refs 3 and 4). In this sense, the Storebreen glacier probably contains a better record of changes in intensities of metal emissions in Europe.

The use of average concentrations of metals in ice layers deposited over a period of several decades or centuries (before 1900) in deriving the data listed by Landy *et al.* is misleading. It assumes that the rate of metal deposition was constant before AD 1900, which is contrary to the available evidence^{3,6}. I have calculated the ranges in the post-1960 to 1800-1900 ratios of reported trace metal concentrations in the ice fields of the Northern Hemisphere (Table A). The maximum and minimum values in reported metal concentrations during 1800-1900 and since 1960 have been used to derive the data listed. Obviously the ranges in the ratios are quite wide and the ratios obtained using my emission data are either within or fall near these ranges. This is 'encouraging' considering that the emission data are only order of magnitude estimates anyway.

The contention by Landy *et al.* that the trace metals in Arctic snow are derived mainly from natural sources is probably not valid. Rahn and his colleagues^{11,12} in fact have recently presented rather convincing evidence that the aerosols at widely dispersed Arctic sites of northern Norway, northern Greenland and Barrow, Alaska are strongly pollution-derived, particularly during the winter time. Before that, Chow and Earl¹³ used the isotope ratio method to show that most of the lead in Greenland glacier is of anthropogenic origin. Ancillary evidence in support of the anthropogenic origin of lead deposited in the snow fields of Northern Hemisphere are given by Settle and Patterson¹⁴. Note that the ratios of the present day deposition rates for Pb, Cd, Cu and Zn at Centrale, Greenland and at Dome C, Antarctica are

65, 16, 16 and 91 respectively¹⁵; the ratios of the Pb, Cd, Cu and Zn concentrations in present day surface snow at the two sites are 6.9, 1.6, 1.6 and 9.4 respectively¹⁵. Presumably, the much higher concentrations and deposition rates at the Arctic site vis-a-vis the Antarctic location is related to the fact that most of the anthropogenic emissions of metals occur in the Northern Hemisphere.

It should be emphasised, however, that thorough studies are needed on the deposition mechanisms and on the factors controlling the elemental composition of polar snow and ice before the metal concentrations in the ice layers can be related quantitatively to the atmospheric metal burdens. On this point, I completely agree with Landy *et al.*

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BOUSTRON COMMENTS—With regard to the above exchange of views I would like to comment on the available polar ice and snow historical records of the variations of the atmospheric concentrations of the five metals (Pb, Cd, Cu, Zn and Ni) discussed in Nriagu's paper¹, in the remote areas of the Northern Hemisphere during the past few centuries. Data such as those published for a temperate glacier in southern Norway² and quoted by Nriagu are not polar data, so that the only genuine polar historical records presently available for the Northern Hemisphere are those obtained in Greenland. Numerous data have been published on the trace metal content of the successive snow and ice layers deposited in the Greenland ice cap during the past few centuries. Unfortunately, many of these data are probably unreliable because they were plagued by severe contamination problems during field collection or (and) during laboratory analysis. It is very important then to estimate which of these data, if any, are reliable: this can be looked at by a detailed examination of the collection and analytical procedures described by the authors, with special reference to the use of high performance clean rooms and to

Table A Ranges in the post-1960 to 1800-1900 ratios of metal concentrations in the Arctic snow fields

| Site | Cd | Cu | Pb | Zn |
|------------------------------------|--------|-------|--------|-------|
| Milcent ⁷ | 1-2 | — | 1-7 | 1-8 |
| Milcent ^{7,8} | <1-10 | — | 3-20 | 1-20 |
| Dye-3 ⁹ | — | — | 9 | — |
| Dye-3 ¹⁰ | 10-600 | 1-100 | — | <1-40 |
| Camp Century ¹⁰ | <1 | 1-7 | — | — |
| Camp Century ⁶ | — | — | <1->12 | — |
| Jotunheimen Mountains ² | <1-68 | — | <1-6 | — |